

**FINAL REPORT FOR NASA GRANT #NAG510425
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**ISOTOPIC AND GEOCHEMICAL INVESTIGATIONS OF
METEORITES**

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Results of Work: 2001-2005.

FINAL REPORT

Overview of work.

The primary goals of our research over the past four years are to constrain the timing of certain early planetary accretion/differentiation events, and to constrain the proportions and provenance of materials involved in these processes. This work was achieved via the analysis and interpretation of long- and short-lived isotope systems, and the study of certain trace elements. Our research targeted these goals primarily via the application of the ^{187}Re - ^{187}Os , ^{190}Pt - ^{186}Os , ^{98}Tc - ^{98}Ru and ^{99}Tc - ^{99}Ru isotopic systems, and the determination/modeling of abundances of the highly siderophile elements (HSE; including Re, Os, Ir, Ru, Pd, Pt, and maybe Tc). The specific events we examined include the segregation and crystallization histories of asteroidal cores, the accretion and metamorphic histories of chondrites and chondrite components, and the accretionary and differentiation histories of Mars and the Moon. The results of the proposed research have: 1) provided us with a better understanding of the metal segregation and crystallization histories/pathways in iron meteorite parent bodies, 2) provided improved constraints on the behavior of HSE during nebular condensation through chondrite formation and subsequent alteration, 3) provided important constraints on the HSE budgets of lunar and Martian mantles, and 4) examined the magnitude of early solar system isotopic heterogeneities in the transition element Mo. The results/progress of six major studies advanced during this funding cycle are reviewed as follows.

1. ^{187}Re - ^{187}Os & ^{190}Pt - ^{186}Os Isotopic and HSE Abundance Studies of Iron Meteorites.

In order to examine the crystallization histories of two major asteroidal cores, the ^{187}Re - ^{187}Os and ^{190}Pt - ^{186}Os systematics of 13 IIAB and 18 IIIAB iron meteorites were analyzed. In contrast to

our previous work, the attainment of low, reproducible Re chemical processing blanks permitted us to make precise Re-Os isotopic measurements for the highest-Ni, lowest-Re IIAB and IIIAB endmembers.

The Re-Os isochron ages for the complete suites of IIAB and IIIAB irons are 4530 ± 50 Ma and 4517 ± 32 Ma, respectively, and are similar to previously reported Re-Os ages for the lower-Ni endmembers of these two groups (e.g. Shen et al., 1996; Smoliar et al., 1996; Chen et al., 2002). The results for both systems are consistent with initial crystallization of both asteroidal cores within several tens of Ma of the origin of the solar system. The results for the most highly-evolved irons in both groups are consistent with, but do not require, complete crystallization during the same time frame as the initiation of crystallization.

The first high-precision Pt-Os isochrons for IIAB (Fig. 1) and IIIAB irons were produced. The Pt-Os isochron ages for the IIAB and IIIAB irons, calculated using the current best estimate of the λ for ^{190}Pt (Begemann et al., 2001), are 4323 ± 80 Ma and 4325 ± 26 Ma respectively. The Re-Os and Pt-Os ages do not overlap within the uncertainties. The younger apparent ages recorded by the Pt-Os system may reflect much later system closure of the most evolved irons from each group, either as a result of Pt diffusion or late-stage crystallization. A more likely possibility is that the decay constant used in the age calculations is incorrect. The slope from the Pt-Os isochron was combined with the age from the Re-Os isochron for the IIIAB irons to calculate a revised value of $1.415 \times 10^{-12} \text{ a}^{-1}$ for the ^{190}Pt decay constant (Cook et al., in review).

The trace element and isotopic compositions of these suites reflect the crystal-liquid fractionation processes involved, and time of crystallization of the asteroidal cores these suites sample. As previously noted for iron meteorite groups and experimental systems, solid metal-liquid metal bulk distribution coefficients (D 's) for both IIAB and IIIAB systems show

Os>Re>>Pt>1, during the initial stages of core crystallization (e.g. Pernicka and Wasson, 1987; Jones and Drake, 1983; Morgan et al., 1995). The latter stages of crystallization for each core are generally characterized by D's for Re>Os>>Pt. The relative partitioning behaviors of these elements are much more complex in the IIIAB core relative to the IIAB core.

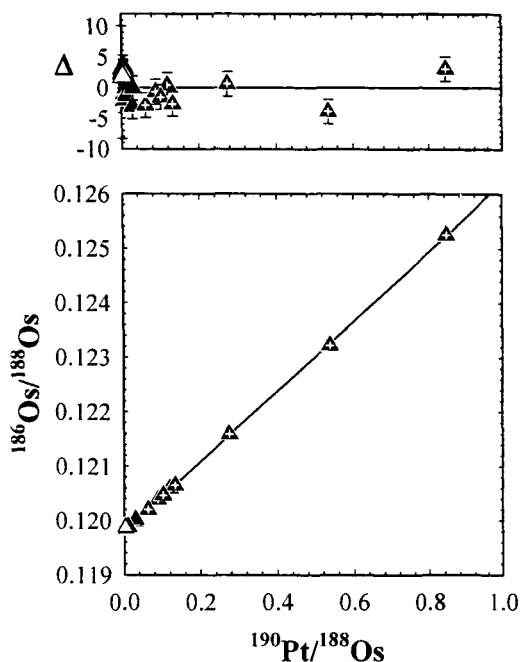


Fig. 1. ^{190}Pt - ^{186}Os isochron for group IIAB iron meteorites (Cook et al., in review). Open triangles-Cape York suite. Solid triangles-low Ni. Triangles with crosses-high Ni. Δ is the per mil deviation from the IIAB isochron.

This work was largely the result of the Masters Thesis work of D.L. Cook (now a Ph.D. candidate at the University of Chicago) and was recently published (Cook et al., 2004). It was done in collaboration with M.F. Horan at the Carnegie Institution of Washington, Department of Terrestrial Magnetism, and J.T. Wasson of UCLA.

2. Analytical work on the ^{98}Tc - ^{98}Ru , ^{99}Tc - ^{99}Ru chronometers.

The short-lived ^{98}Tc - ^{98}Ru ($t_{1/2}$ between 4 and ~ 10 Ma), ^{99}Tc - ^{99}Ru ($t_{1/2} = 0.21$ Ma) chronometers were studied because they may yield important information on the relative chronology of processes that affected metal phases in the early solar system. These include volatility-related processes, such as condensation and evaporation, metal-silicate segregation in asteroids, early core crystallization and aqueous alteration. Furthermore, the proof of extant ^{99}Tc in the solar system would also constrain the time interval between production of the nuclide and injection of s-process isotopes into the protosolar cloud. Previous studies of the Tc-Ru systems reported positive deviations for ^{98}Ru in iron meteorites relative to a terrestrial standard, a small positive deviation for ^{99}Ru in chondritic metal, but normal compositions for calcium-aluminum-rich inclusions (CAIs) from chondrites.

Our Ru isotopic data for group IIAB irons (Negrillos, Bennett County, Coahuila, Filomena, Old Woman) and IIIAB irons (Casas Grandes, Costilla Peak), and the chondrites Allende (CV3) and Allegan (H5) indicate that the abundances of ^{98}Ru and ^{99}Ru in these meteorites overlap with the terrestrial values at the ± 0.8 and ± 0.3 ϵ levels (parts in 10,000), respectively (Figs. 2-3). Lack of heterogeneities of ^{98}Ru and ^{99}Ru among the iron meteorites is not surprising in light of a recent experimental study that shows that crystal-liquid fractionation of metal changes the Tc/Ru (and Tc/Mo) ratio by substantially less than a factor of 2 (Lazar et al., in review). Using these constraints and solar system initial abundances of ^{99}Tc derived from astrophysical models, evolution models for the ^{99}Tc - ^{99}Ru chronometer can produce barely resolvable deviations in ^{99}Ru (0.3 ϵ) only if these fractionation events occurred during collapse of the molecular cloud that formed our solar system (< 10 ka, Becker and Walker, 2003). Absence of ^{98}Ru deviations indicates $^{98}\text{Tc}/^{96}\text{Ru}_{\odot i} < 2 \times 10^{-5}$. Ratios of the stable isotopes of Ru also overlap within error with the terrestrial values at better than 1 ϵ unit (Becker and

Walker, 2003). However, it should be noted that the data for Allende are also consistent with a ~ 1 ϵ deficit of pure s -process ^{100}Ru , relative to p - and r -process isotopes, as observed in another recent Ru isotopic study (Chen et al., 2003). The latter study also reported 0.3-1.0 ϵ deficits of ^{100}Ru in differentiated meteorites.

Some results of our study were published in Becker and Walker (2003). A paper detailing analytical issues and comparing N-TIMS with multi-collector ICP-MS data has also been published (Becker et al., 2002).

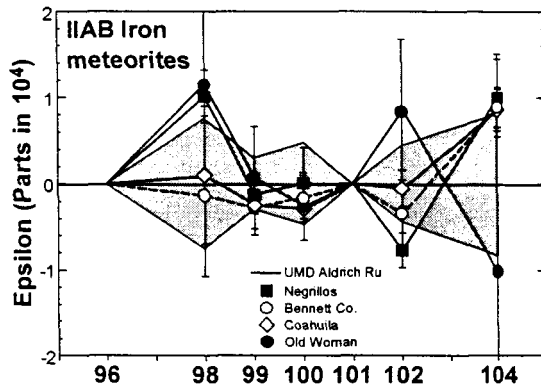


Fig. 2. Ru isotopic composition of IIAB iron meteorites. Data are means of multiple analyses. Uncertainties are $2\sigma_m$. Grey area indicates $\pm 2\sigma_m$ uncertainty of multiple ($n = 4-6$) standard runs.

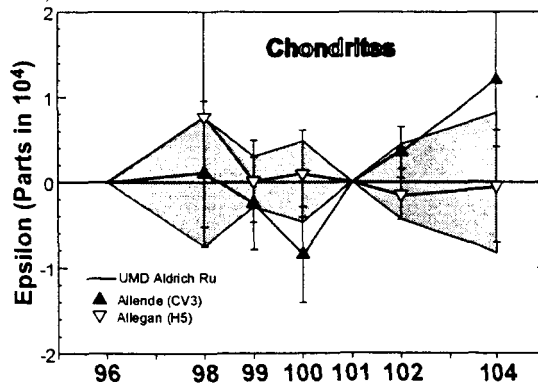


Fig. 3. Ru isotopic composition of the chondrites Allende and Allegan. Data are

individual analyses. Uncertainties are $2\sigma_m$ (in-run).

3. Scale of isotopic heterogeneity between inner solar system bodies.

Recent Mo isotopic studies on meteorites reported evidence for differences in isotopic compositions for whole rocks of some primitive (Yin et al., 2002; Dauphas et al., 2002a, b) and differentiated meteorites (Qi-Lu and Masuda, 2000; Dauphas et al., 2002a), relative to terrestrial materials. Relative enrichments of r - and p -process isotopes of up to 3-4 ϵ units (ϵ unit = parts in 10^4) over s -process dominated isotopes are the most prominent features. Certain types of presolar grains show enrichments in s -process isotopes (Nicolussi et al., 1998). If the reported variability in r - and p -process isotope enrichments reflects the true isotopic characteristics of the whole rocks, the implications are quite profound. It would suggest the presence of large scale Mo isotopic heterogeneity within the solar accretion disk with likely collateral effects for other elements. Because similar effects are not apparent from our Ru isotope work, nor from Zr isotopic studies of meteorites (Schönbächler et al., 2002), we examined the Mo isotopic composition of iron meteorites, chondrites and CAIs using negative thermal ionization mass spectrometry.

With the exception of ^{100}Mo , the Mo isotopic composition of the bulk meteorites and CAIs we analyzed overlap with terrestrial Mo within ± 1 ϵ unit, when normalized to $^{92}\text{Mo}/^{98}\text{Mo}$ (Figs. 4-5). ^{100}Mo is the only Mo isotope outside the mass range used for normalization, and hence, may yield the least precise results. Our results on IIAB iron meteorites and the Allende whole rock show no evidence for enrichments in p - or r -process isotopes, relative to pure s -process ^{96}Mo . In contrast, two CAIs from Allende show patterns that bear a similarity to the whole rock patterns for Allende reported by Yin et al. (2002) and Dauphas et al. (2002a), and patterns of

Allende leachates and residues (Dauphas et al., 2002b).

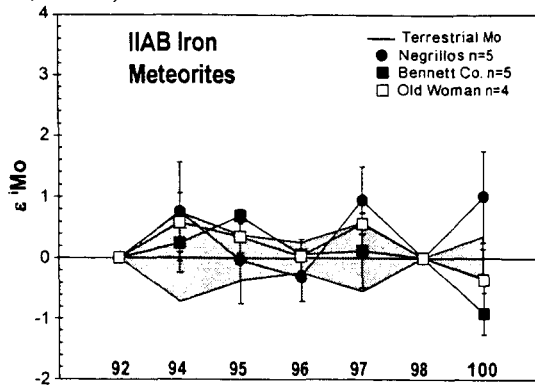


Fig. 4. Isotopic composition of Mo in IIAB iron meteorites, normalized to terrestrial Mo measured at UMD and displayed as deviations in ϵ units. Data are means of multiple analyses. Uncertainties are $2\sigma_m$.

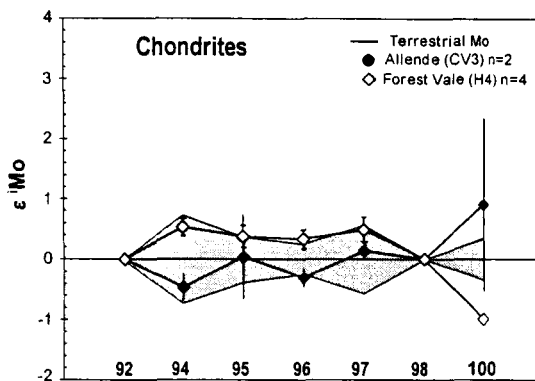


Fig. 5. Mo isotopic composition of chondrites Allende (CV3) and Forest Vale (H4). Data are means of multiple analyses. Uncertainties are $2\sigma_m$.

The reason why some research groups find isotopic heterogeneity in the same groups of iron meteorites (Qi-Lu and Masuda, 2000; Dauphas et al., 2002a), and others don't (Yin et al., 2002, Becker and Walker, 2002) remains unresolved. The different results obtained for whole rocks of Allende by different groups could simply reflect a sample digestion issue (teflon beakers vs. Carius tube). Alternatively, it

could reflect heterogeneity of Allende on the cm-scale, since it appears that some of its components, such as CAIs, show isotopic heterogeneity (Fig. 6, see also Yin et al., 2002; Dauphas et al., 2002b).

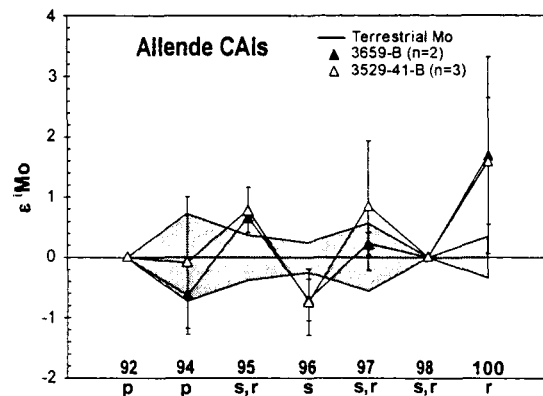


Fig. 6. Mo isotopic composition of CAIs from Allende. Data are means of multiple analyses. Uncertainties are $2\sigma_m$.

Because of these unresolved issues it is difficult to assess whether or not the existing Mo isotope data indicate large-scale isotopic heterogeneity in the inner solar system. While our new Mo data are consistent with existing Zr isotopic data, new Ru isotopic data by Chen et al. (2003) may hint at a small deficit of the pure s-process ^{100}Ru isotope in some differentiated meteorites and in Allende. This in turn *may* indicate large-scale isotopic heterogeneity in the inner solar system.

4. ^{187}Re - ^{187}Os and HSE Systematics of Chondrites

During this funding period we completed long-term studies of the ^{187}Re - ^{187}Os isotopic, and HSE elemental systematics of bulk chondrites (Walker et al., 2002a; Horan et al., 2003). For the Re-Os work, a suite of 47 carbonaceous, enstatite and ordinary chondrites were examined. We discovered significant differences in the $^{187}\text{Re}/^{188}\text{Os}$ and $^{187}\text{Os}/^{188}\text{Os}$ ratios of carbonaceous chondrites compared with ordinary and enstatite chondrites. The

average $^{187}\text{Re}/^{188}\text{Os}$ for carbonaceous chondrites is 0.392 ± 0.015 (excluding the anomalous CK chondrite, Karoonda), compared to 0.422 ± 0.025 and 0.421 ± 0.013 for ordinary and enstatite chondrites (1σ standard deviations). These ratios recast into elemental Re/Os ratios are: 0.0814 ± 0.0031 , 0.0876 ± 0.0052 and 0.0874 ± 0.0027 , respectively. Correspondingly, the $^{187}\text{Os}/^{188}\text{Os}$ ratios of carbonaceous chondrites average 0.1262 ± 0.0006 (excluding Karoonda), while ordinary and enstatite chondrites average 0.1283 ± 0.0017 and 0.1281 ± 0.0004 , respectively (1σ standard deviations). The results indicate that the Re/Os ratios of meteorites within each group are, in general, quite uniform. The minimal overlap between the isotopic compositions of ordinary and enstatite chondrites versus carbonaceous chondrites indicates long-term differences in Re/Os for these materials, most likely reflecting chemical fractionation early in solar system history.

A majority of the chondrites do not plot within analytical uncertainties of a 4.56 Ga reference isochron (Fig. 7a-b). Most of the deviations from the isochron are consistent with minor, relatively-recent redistribution of Re, and/or Os on a scale of mm to cm. Some instances of the redistribution may be attributed to terrestrial weathering, others are most likely the result of aqueous alteration or shock events on the parent body within the past 2-3 Ga.

The $^{187}\text{Os}/^{188}\text{Os}$ ratio of Earth's primitive upper mantle has been estimated to be 0.1296 ± 8 (Meisel et al., 2001). If this composition was set via addition of a late veneer of planetesimals following core formation, the composition suggests the veneer was dominated by materials that had Re/Os ratios most similar to ordinary and enstatite chondrites.

In order to examine the behavior of the HSE in chondrites further, we generated high-precision HSE patterns for a broad sampling of ordinary, enstatite and carbonaceous chondrites. This work was done in collaboration with M.F. Horan at the Carnegie Institution of Washington,

Department of Terrestrial Magnetism. As with the Re/Os ratio, there are significant differences in relative HSE abundances between the 3 main chondrite groups (Horan et al., 2003). Of greatest note is the distinctly higher average Pd/Ir present in some enstatite chondrites relative to ordinary and carbonaceous chondrites (Fig. 8). The fractionations of the HSE are presumably the result of both high temperature nebular and secondary processes, such as impact events.

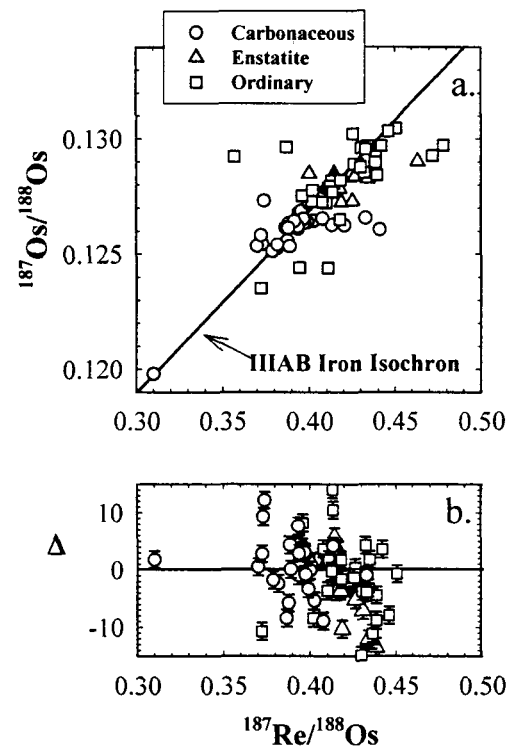


Fig. 7. a. $^{187}\text{Re}/^{188}\text{Os}$ versus $^{187}\text{Os}/^{188}\text{Os}$ for all bulk chondrites analyzed by this study. Shown for reference is the 4.557 Ga group IIIAB iron meteorite isochron reported by Smoliar et al. (1996). **b.** $^{187}\text{Re}/^{188}\text{Os}$ versus Δ for all bulk chondrites analyzed by this study. Δ is the per mil deviation of a sample from the IIIAB iron isochron of Smoliar et al. (1996). These results were published in Walker et al. (2002a).

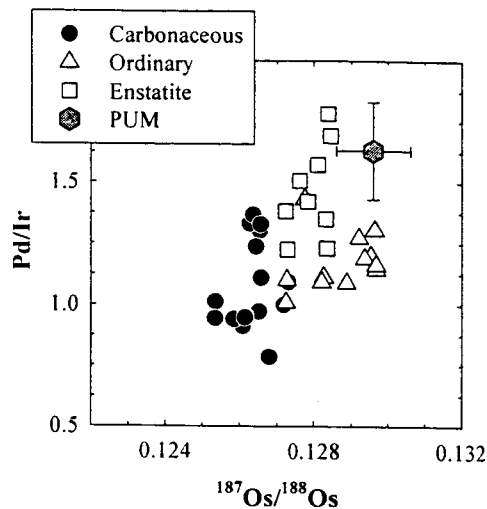


Fig. 8. Bulk chondrite Pd/Ir ratios versus $^{187}\text{Os}/^{188}\text{Os}$. Also shown for comparison is the current best estimate for the Earth's primitive upper mantle (PUM) from Meisel et al. (2001) and Morgan et al. (2001).

As a means of examining the distributions of HSE in chondrites and determine when the HSE were mobile, we have also begun analysis of chondrite components including: chondrules, calcium aluminum inclusions (CAIs), matrix and sulfides (Becker et al., 2001; Walker et al., 2002). Most materials examined to date were from the Allende meteorite. Most CAIs plot very close to the iron meteorite isochrons and display a modest range in $^{187}\text{Os}/^{188}\text{Os}$ (Fig. 9). CAIs, with highly fractionated REE patterns (Group II), display a larger range in Re/Os and $^{187}\text{Os}/^{188}\text{Os}$ than the other CAIs. If representative of early condensates, the limited distribution of $^{187}\text{Os}/^{188}\text{Os}$ for most CAIs, except those with Group II REE patterns, indicate a solar system value for $^{187}\text{Os}/^{188}\text{Os}$ of approximately 0.127. Our work on chondrites has resulted in the publication of three papers during this funding cycle (Becker et al., 2001; Walker et al., 2002a; Horan et al., 2003).

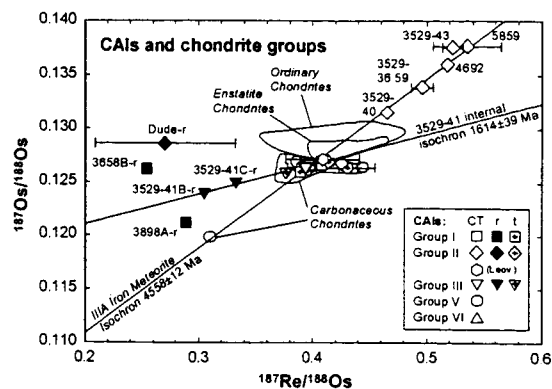


Fig. 9. Isochron plot of CAI data for Allende and Leoville. CT = Carius tube data; r = residues from Carius tube digestions; t = combined CT and r data (see Becker et al., 2001 for details). Fields for whole rock samples of carbonaceous, ordinary and enstatite chondrites are shown for comparison. The IIIA iron meteorite isochron from Smoliar et al. (1996) is shown for comparison.

5. Re-Os isotopic studies of SNC meteorites.

Determining the absolute and relative abundances of HSE in the Martian mantle are of great importance. As an example, the abundances of Os we have measured in most SNC meteorites lie within, but at the low end of the range of data defined by terrestrial volcanic rocks, and within the range of terrestrial cumulate rocks (e.g. pyroxenites) (Fig. 10). This may suggest that the abundances of the HSE in the Martian mantle are similar to, or only a factor of 2-5 lower than in the terrestrial mantle (Walker et al., 2002b). This, in turn, may indicate a late accretionary period similar to that experienced by Earth, that buffered the abundances of the HSE at much higher levels than what remained in the silicate Mars immediately subsequent to core formation. Similar conclusions were reached from the abundances of other HSE, most notably Pt (Jones et al., 2003).

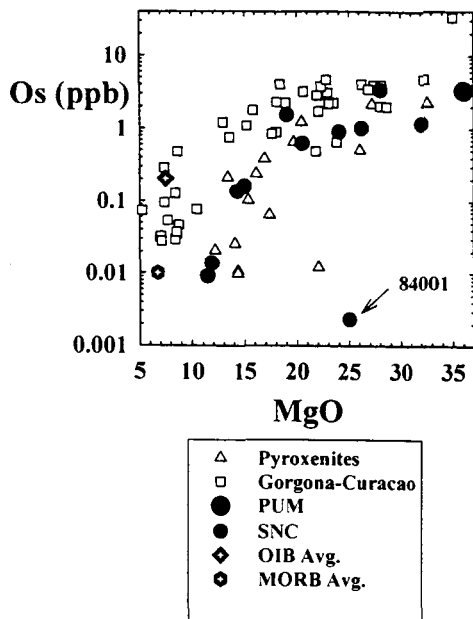


Fig. 10. Abundances of MgO (wt. %) plotted versus Os (in ppb) for SNC meteorites. SNCs define a J-shaped pattern similar to terrestrial rocks from the Caribbean plateau (Gorgona-Curacao) and terrestrial pyroxenites. Only ALH 84001 plots well below the trend. From Walker et al. (2002b).

Variations in the short-lived systems of ^{182}Hf - ^{182}W and ^{146}Sm - ^{142}Nd in the SNC meteorites indicate an early isolation of, and subsequent inefficient mixing between, mantle reservoirs in Mars (Lee and Halliday, 1997; Harper et al., 1995). Thus, early differentiation histories may be preserved in other isotopic systems in these meteorites as well. The Re-Os isotopic systematics of Martian meteorites can provide valuable information on what chemical processes affected the Martian mantle, particularly with regard to the relative abundances of highly siderophile elements (HSE), including core formation, late veneer addition, and early melting. Towards that end we are studying the Re-Os isotopic systematics of the SNC meteorites to determine which, if any, of these processes affected the HSE budget of the Martian

mantle and the implications for the early differentiation history of Mars.

We have now examined ten SNC meteorites for Re-Os isotopic compositions. The best estimates of the initial γ_{Os} values for SNC meteorites indicate a long-term Re depletion history for the NC group, with a present day γ_{Os} value extrapolated to be approximately -3 ± 2 . In contrast the S-group meteorites were derived from a source with a long-term, slightly suprachondritic Re/Os. This source has evolved to a higher γ_{Os} ($+5 \pm 2$) than the present-day primitive upper mantle of Earth ($+1.6 \pm 0.8$). The source that generated EETA 79001 had an Os isotopic composition consistent with chondritic Re-Os evolution. Of note, the variations in Re/Os of the two apparently endmember mantle sources appear to have been set during the earliest differentiation history of Mars. This is indicated by the correlations between initial γ_{Os} , and $\epsilon_{182\text{W}}$, and $\epsilon_{142\text{Nd}}$ (Fig. 11).

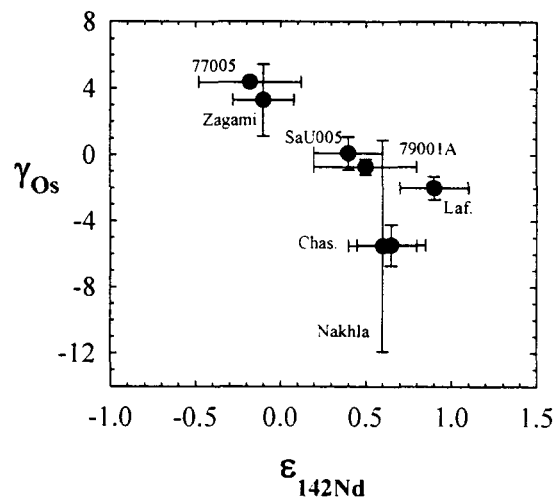


Fig. 11. γ_{Os} versus $\epsilon_{142\text{Nd}}$ plot for SNC meteorites. Os data are from Brandon et al. (2000) and Walker et al. (2002b). Nd data are from Harper et al. (1995) and L. Nyquist (pers. comm.).

Although the SNC meteorites crystallized at different times, extrapolations to any one time during the last 2 Ga would not cause major shifts in the relations,

because of the presumed, nearly chondritic Re/Os of the sources. The negative correlation between γ_{Os} and ϵ_{142Nd} is especially interesting because both systems likely reflect early melt depletion (Nd and Re removal). Conversely, the S meteorites are consistent with derivation from a melt enriched (Re and Nd addition) reservoir, possibly even a relict of an early magma ocean. EETA 79001 (lithology A) is either derived from a mixed source, or alternatively, a primitive, un-differentiated source. Initial results of this work were published in Brandon et al. (2000).

6. HSE Systematics of the Lunar Interior.

As with Mars, the absolute and relative abundances of the HSE present in the lunar mantle may provide unique insights to the formation and late accretionary history of not only the Moon, but also Earth. Determining the HSE content of the lunar mantle, however, has proven difficult, because no *bona fide* mantle rocks have been collected from the moon. Primitive materials, such as picritic glasses are potentially the most diagnostic materials we have for constraining HSE abundances in the lunar interior (e.g. Ringwood, 1992; Richter et al., 2000). Consequently, we have begun to determine Os isotopic compositions and Re, Os, Ir, Ru, Pt and Pd concentration data for green and orange glasses. This work is being done in collaboration with M.F. Horan at the Carnegie Institution of Washington, Department of Terrestrial Magnetism, and C. Shearer and J. Papike at the University of New Mexico. As with previous studies utilizing neutron activation analysis, we are examining different size fractions of the glass spherules to assess the role of surface condensation in the generation of the HSE abundances.

Absolute and relative abundances of HSE in green and orange glasses are highly variable (Fig. 12). Some fractions of orange glass we have examined are characterized by highly fractionated HSE patterns with major depletions in Os and Ir. In contrast, some fractions (particularly of green glass) are relatively flat, indicating that the HSE

abundances of these glasses are dominated by a meteoritic contaminant. Mixing calculations indicate that even very minor amounts of a meteoritic contaminant have strongly affected the HSE concentrations and Os isotopic compositions of most glasses (Fig. 13).

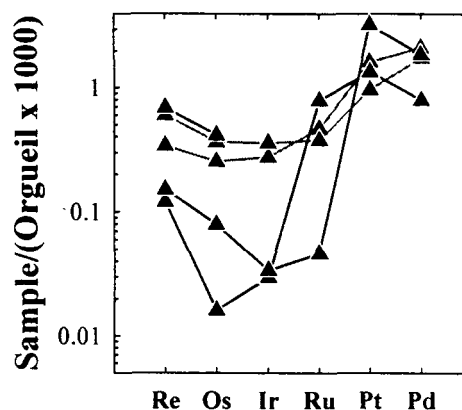


Figure 12. CI chondrite normalized HSE plot for orange glasses 74220 and 74001. Note that one of the glass fractions has quite low Os and a highly fractionated pattern.

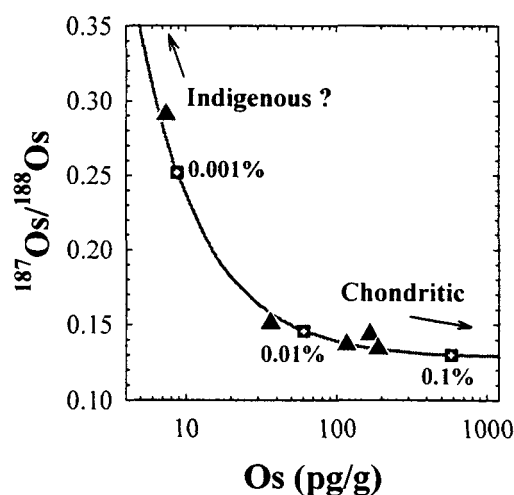


Figure 13. Mixing calculations suggest that all orange glasses analyzed contain some proportion of a chondritic contaminant. One sample, with quite radiogenic $^{187}Os/^{188}Os$ may approach the composition of the indigenous orange glass component.

In those samples with the most fractionated HSE patterns, Os and Ir concentrations are quite low, and even Pt and Pd concentrations are a factor of 6-8 lower than abundances for terrestrial volcanic rocks with similar MgO. This may indicate that the lunar mantle source of at least the orange glasses had bulk HSE abundances substantially lower than the terrestrial (or evidently the Martian) mantle. This, in turn, may suggest that a significant portion of the late veneer added to the Earth-Moon system occurred subsequent to the formation of the lunar crust at 4.2-4.5 Ga. Results of this work were recently published (Walker et al., 2004).

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August 8, 2005

Dr. D. Lindstrom
NASA Headquarters, Code SR
Washington, D. C. 20546

Dear Dave,

Please find enclosed a copy of the final report for my NASA Cosmochemistry grant NAG510425, which was active from 2001 to 2005. Please note that there is a list of publications resulting from this grant on the last page. If you have any questions regarding this research, please feel free to contact me at 301-405-4089.

Thanks for the support.

Sincerely,

Richard J. Walker
Professor of Geology

cc. Brad Poston



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August 8, 2005

Mr. Brad Poston
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Greenbelt, MD 20771

Dear Mr. Poston,

Please find enclosed a copy of the final report for my NASA Cosmochemistry grant NAG510425, which was active from 2001 to 2005. Please note that there is a list of publications resulting from this grant on the last page. If you have any questions regarding this research, please feel free to contact me at 301-405-4089.

Thanks for the support.

Sincerely,

Richard J. Walker
Professor of Geology

c.c. D. Lindstrom